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14. ABSTRACT

Work supported by this grant led to major theoretical and experimental advances in the physics of sodium (Na) guide stars for use in adaptive optics. We developed a powerful and realistic new theory of the effect of velocity-changing and spin changing collisions on optically pumped atoms at low pressures, similar to those of Na guide star atoms at altitude of about 100 km in the earth's atmosphere. Particularly useful was the development of new "cusp kernels" to describe the combined effects of strong velocity changing collisions acting simultaneously with spin precession in geomagnetic fields and spin-exchange collisions with oxygen molecules and dissociated oxygen atoms in the upper atmosphere. We implemented laboratory experiments that mimicked as closely as possible the collisional conditions of the upper atmosphere, given the unavoidable need for constraining cell walls. We developed a new, temperature-insensitive way to use Faraday rotation to lock a tunable diode laser to an arbitrary frequency in the absorption spectrum of an atomic resonance line.

15. SUBJECT TERMS

Laser frequency stability, adaptive optics, optical pumping, spin relaxation, velocity-changing collisions, collision kernels.

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FINAL TECHNICAL REPORT

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PHYSICS OF OPTICALLY PUMPED ALKALI-METAL ATOMS

Submitted August 31, 2013
by
William Happer
Principal Investigator

1. Overview

The research supported by this grant was focused on the fundamental physics and applications of spin polarized atoms. The principal investigator, William Happer, is very grateful for the support he has received from AFOSR over most of his career in physics. At the end of the academic year, 2013-1014, he will retire from teaching at Princeton University. He plans to continue activities in physics in an emeritus status, but without the graduate students and post docs that have meant so much to him. He will devote as much time as possible to the physics of the Earth's greenhouse effect. How atoms and molecules emit and absorb radiation in gases with pressure ranges and temperature ranges comparable to those of the earth's atmosphere has been a central focus of Happer's AFOSR-supported research, and will be an great advantage for work on greenhouse physics. Of particular interest is how collisions of CO_2 molecules with the nitrogen and oxygen molecules of the air affect the way CO_2 absorbs and emits radiation. Incorrect modeling of collisional line broadening is one of several possible reasons that climate models have so greatly exaggerated the warming from more CO_2. There has been basically no warming at all for about 15 years, contrary to model predictions of about 0.3 C temperature increase for the increase of atmospheric CO_2 from 370 ppm to 400 ppm. Ultimately, the truth about collisional effects will have to be resolved by incisive experiments. Theory alone is not yet sufficiently quantitative.

To give the flavor of the work supported by this grant, in the next section we will summarize research directions of particular interest to the principal investigator, with a special focus on the more theoretical activities. More details about the extensive experimental work supported by the grant can be found in the publications listed at the end of this report. Also listed there are students and other researchers who received full or partial support from the grant.

2. Some Major Accomplishments

The book, Optically Pumped Atoms. The principal investigator, William Happer, and two former students, Thad Walker, now a professor of physics at the University of Wisconsin, and Dr. Yuan-Yu Jau now a member of the professional staff at Sandia National Laboratory, wrote a book "Optically Pumped Atoms," published by Wiley in 2010

[8] which summarizes many of the powerful new theoretical methods that have been developed for modeling the evolution of optically-pumped, spin-polarized atoms in the presence of optical or microwave/radiofrequency radiation, static magnetic fields, and various types of collision processes. The book shows how to treat real atoms, with many sublevels in the ground state and optically excited state, with the same degree of rigor as the idealized "two-level atom," which is widely discussed because of its simplicity, but which is of limited use for modeling quantitative details for real atoms.

A particularly useful way to describe spin-polarized atoms is with a density matrix ρ. The book explains how the density matrix can be represented by a column vector $|\rho)$ in Liouville space, with a time-evolution equation

$$\frac{d}{dt}|\rho) = -G|\rho). \tag{1}$$

Often the the evolution matrix G is independent of $|\rho)$. For systems with oscillation perturbations like the effects of microwave radiation or laser light, a generalized "rotating coordinate system" can often be found for which G can be taken to be time independent. So (1), often called the "master equation" is often a first-order, vector linear equation with time-independent matrix G of coefficients. The challenge is that the column vector $|\rho)$ representing the density-matrix ρ has a very large dimension, 256 in the case of ground-state ^{133}Cs atoms, and the dimension can easily exceed 1000 if elements of the excited states are included. As stressed in the book, modern mathematical software like MATLAB is well adapted to calculations in such high-dimensional spaces.

Unlike the analogous, Schroedinger equation

$$\frac{d}{dt}|\psi\rangle = -i\hbar H|\psi\rangle. \tag{2}$$

which evolves under the influence of Hamiltonian matrix H which is Hermitian, $H^\dagger = H$, the damping operator G of the master equation (1) is neither Hermitian nor "normal," that is

$$G^\dagger \neq G, \quad \text{and} \quad G\,G^\dagger \neq G^\dagger G. \tag{3}$$

We assume that the evolution matrix G of (1) has right (column or ket) eigenvectors $|g)$ and eigenvalues g defined by

$$G|g) = g|g). \tag{4}$$

Since G is not Hermitian the eigenvalues g of G need not be real,

$$g \neq g^*. \tag{5}$$

and the left (row or bra) eigenvectors, which we denote by a double left parenthesis as $(\!(g|$ and define by

$$(\!(g|G = (\!(g|g, \quad \text{with} \quad (\!(g| \neq |g)^\dagger. \tag{6}$$

need not be Hermitian conjugates $|g)^\dagger$ of the right the eigenvector $|g)$, defined by (4). The situation is reminiscent of x-ray crystallography, where it is customary to introduce

reciprocal lattice vectors (left eigenvectors, row eigenvectors) that differ from the lattice vectors (right eigenvectors, column eigenvectors) to efficiently analyze diffraction patterns.

Systematically building on the simple ideas noted above the, book shows how to use modern mathematical software like MATLAB to efficiently solve very complicated optical pumping problems where the density matrix has hundreds of elements. The notation of the book was chosen to facilitate computer coding, so that a symbolic equation involving vectors or matrices looks almost the same when converted to a line of code in MATLAB. The book also contains many software packages for optical pumping that can be downloaded from a permanent website at the University of Wisconsin for use in specialized problems.

Velocity changing collisions of spin-polarized atoms. One of the significant results of AFOSR research was the principal investigator's invention of the the "sodium guide star," for sensing and correcting for the effects of optical turbulence on propagation of lasers, for sharpening ground based imaging of space objects, and for sharpening the resolution of large, ground-based astronomical telescopes. The Na guidestar concept was heavily classified throughout the 1980's, when extensive experimental work led by Dr. Bob Fugate at Kirtland Air Force Base demonstrated its usefulness. At altitudes between about 90 and 100 km, there are free Na atoms in sufficient abundance to backscatter a significant fraction of laser light tuned to the center of the Na resonance line. The Na atoms are produced by the vaporization of micro-meteoroids that constantly bombard the earth. The altitude of the Na layer is where the micro-meteoroids get hot enough from "reentry" heating to volatilize most of their sodium atoms or to "burn up" entirely. All large modern telescopes now include a sodium guidestar system, which consists of a laser located at the observatory that can be tuned to the 590 nm D2 line of sodium atoms and directed toward the star or galaxy of interest. The backscattered laser light from these atoms can serves as an artificial bright star to probe the atmospheric turbulence and close feedback loops to correct for the degradation in spatial resolution or " seeing" that is caused by the turbulence.

Although Na guidestar systems work well in astronomical applications, there are no return photons to spare. One can use physics to increase the backscatter without using more powerful lasers. One of the most useful ways to increase Na guidestar signals is to optically pump atoms into spin sublevels with large backscatter cross sections. The physics related to this pumping differs quite substantially from that encountered in most laboratory experiments on optical pumping, which are normally done at much larger gas pressures, as in the case of gas-cell optical pumping, or done with no background gas at all, as in the case of optically trapped, cold atoms. At the altitude of the sodium layer, the the total number density the atmospheric nitrogen and oxygen molecules and the relatively small number of dissociated oxygen atoms is only about $N = 10^{14}$ cm^{-3}. The rate coefficients for collisions of the Na atoms on the residual gas molecules and atoms are of order $\langle v\sigma \rangle = 10^{-10}$ cm^3 s^{-1}, so collision rate is only $\gamma = N\langle \bar{v}\sigma \rangle = 10^4$ s^{-1}, much slower than the natural radiative decay rate of excited Na atoms, $1/\tau = 6 \times 10^7$ s^{-1} for the radiative lifetime $\tau = 16$ ns of the first excited state of Na.

Because of the slow collision rate, the overall width of the optical absorption cross section will be almost unaffected by collision broadening and will be almost completely determined by hyperfine structure, natural radiative broadening and Doppler broadening.

When pumped by nearly monochromatic ground-based lasers, only those Na atoms moving with "range velocities" v that Doppler shift them into resonance will absorb laser light. To analyze Na guidestar atoms, one must therefore keep track not only of the spin-state of the atoms, but also of the velocity state [10]. Denote an element of the atomic density matrix for atoms in the ground state by ρ_r, where r denotes the velocity-independent "relaxation rate" of the atom. To include both population and coherent components of the density matrix, r is taken to be a complex number with the real part representing relaxation and the imaginary part representing the spin-precession frequency. A single collision can cause a large change in velocity, so it is not possible to describe the evolution of ρ_r with a partial differential equation. Instead the evolution is described by the integral equation [10],

$$\frac{\partial}{\partial t}\rho_r(v,t) = -\int_{-\infty}^{\infty}(v|K_r|v')\rho_r(v',t)dv' + S_r(v). \tag{7}$$

Here $(v|K_r|v')$ is the kernel of the integral transform that describes velocity-changing collisions, and $S_r(v)$ is the rate of creation of the density matrix element ρ_r with relaxation rate r and velocity v. For simplicity, we have taken S_r to be independent of time, but it is straightforward to consider time-dependent rates. The dimensionless velocity v is measured in units for which a Maxwellian distribution would be e^{-v^2}.

For weak collisions, each of which changes the velocity by a small amount, the integral equation (7) can be well approximated by a differential equation,

$$\frac{\partial}{\partial t}\rho_r = -\left(-\frac{1}{2}\frac{\partial^2}{\partial v^2} - \frac{\partial}{\partial v}v + r\right)\rho_r + S_r. \tag{8}$$

The differential equation (8) represents forced diffusion toward a steady-state, Maxwellian velocity distribution, e^{-v^2}, since

$$\left(\frac{1}{2}\frac{\partial^2}{\partial v^2} + \frac{\partial}{\partial v}v\right)e^{-v^2} = 0. \tag{9}$$

For steady state, when $\partial\rho_r/\partial t = 0$, the formal solution to (8) is

$$\rho_r(v,t) = \int_{-\infty}^{\infty}(v|G_r|v')S_r(v')dv'. \tag{10}$$

The Green's function for the velocity-diffusion equation (8) is the solution to

$$\left(-\frac{1}{2}\frac{\partial^2}{\partial v^2} - \frac{\partial}{\partial v}v + r\right)(v|G_r|v') = \delta(v-v'). \tag{11}$$

In an important advance, Happer and Morgan [10] showed that one can write the Green's function defined by (11) in the closed form

$$(v|G_r|v') = \frac{2^r}{\sqrt{\pi}}\Gamma(r)e^{v'^2}R_r(-v_<)R_r(v_>). \tag{12}$$

where $v_<$ is the lesser of v and v' and $v_>$ is the greater. $\Gamma(r)$ is Euler's gamma function for the complex argument r. The "right" functions R_r are defined, except for normalization, by the differential equation

$$\left(-\frac{1}{2}\frac{\partial^2}{\partial v^2} - \frac{\partial}{\partial v}v + r\right) R_r(v) = 0, \tag{13}$$

with the boundary condition

$$\left(-\frac{1}{2}\frac{d}{dv} - v\right) R_r(v) \to 0, \quad \text{as} \quad v \to \infty. \tag{14}$$

The right function is entire (that is, it has no singularities for finite values of v in the complex v plane). A convenient expression for R_r, is the power series [10]

$$R_r(v) = \sum_{n=0}^{\infty} \frac{\sqrt{\pi}(-v)^n}{n! 2^{r-n} \Gamma(\frac{1+r-n}{2})}. \tag{15}$$

Cusp kernels. There have been very few laboratory studies of optical pumping under conditions like those of optically excited Na atoms at an altitude of about 100 km. The residual atmospheric pressure, on the order of millitorr, is very small by the standards of laboratory gas cell experiments, but it is still much higher than the pressures for work with laser cooled and trapped atoms. To analyze the few experiments that have been done, one assumes that a polarized element of the density matrix, $\rho(v,t)$, evolves according to the integral equation,

$$\frac{\partial}{\partial t}\rho(v,t) = -(\gamma_{sd} + \gamma_{vd})\rho(v,t) + \gamma_{vd} \int W(v,v')\rho(v',t)dv' + P(v,t). \tag{16}$$

The first term on the right of (16) represents losses of the atomic polarization at at velocity v by spin-damping collisions at a velocity independent rate γ_{sd} and losses due to transfer to other velocity groups at the velocity-damping rate γ_{vd}. The second term on the right of (16) represents the collisional transfer of polarization from an initial velocity v' to polarization observed at the velocity v. The third term represents creation of polarization by optical pumping at the rate $P(v,t)$. Eq. (16) is of the same form as (7).

The most widely used model for the collision kernel $W(v,v')$ of (16) has been the Keilson-Storer (KS) kernel [15]

$$W_a(v,v') = \frac{e^{-(v-av')^2/b^2}}{b\sqrt{\pi}}, \tag{17}$$

The KS kernel has a single, real "memory parameter" a, with $0 \le a \le 1$, and a corresponding width $b = \sqrt{1-a^2}$. From inspection of (17) one sees that the KS kernel assumes that on average, a single collision converts atoms with a velocity v' into atoms with a Gaussian distribution of velocities v and a variance b^2. The new distribution is centered

on the velocity av', that is, the atoms "remember" a fraction a of the their pre-collision velocity v'.

The KS distribution appears to have been first introduced to physics in 1891 by Raleigh [10] in connection with his studies of velocity distributions produced by weak collisions. It turns out to the impulse response function of the differential equation (8) at the time $\tau = -\ln a/r$, that is, it is the distribution of velocities v at time $\tau > 0$ that would have evolved as described by (8) from the distribution $\delta(v-u)$ at initial time $\tau = 0$.

The collision kernel $W(v,v')$ of (16) is the probability that a collison transfers an atom with an initial velocity v' to a final velocity between v and $v+dv$. It must therefore satisfy the normalization condition

$$\int_{-\infty}^{\infty} dv W(v,v') = 1. \tag{18}$$

Collisions should also transform a Maxwellian distribution, $e^{-v'^2}$, of initial velocities into the same Maxwellian distribution of final velocities, so a physically acceptable kernel must satisfy the constraint

$$\int_{-\infty}^{\infty} W(v,v') e^{-v'^2} dv' = e^{-v^2}. \tag{19}$$

The KS kernel (17) satisfies (18) and (19), and it has the advantage of a simple analytic form. The basic problem with the KS kernel is that it does not resemble kernels deduced from experimental measurements. When plotted against the final velocity v, observed kernels have a sharp cusp centered on the initial velocity v'. They do not look at all like the Gaussian KS kernel. The origin of the cusp is the large number of weak collisions that correspond to large impact parameters.

Working with students Robert Marsland, Bart McGuyer and Ben Olsen, Happer developed a powerful new "cusp kernel" [15] that looks very much like experimentally determined kernels, satisfies important physical constraints (18) and (19), and has a simple analytic form determined with a single sharpness parameter s, that is analogous to the memory parameter a of the KS kernel. The cusp kernel can be written as a superposition of KS kernels with a continuous distribution of memory parameters a.

$$C_s(v,v') = s\int_0^1 W_a(v,v') a^{s-1} da. \tag{20}$$

For large sharpnesses $s \gg 1$, the cusp kernel is dominated by realatively sharp KS kernels of memory parameters a close to 1. It turns out [15] that the cusp kernel has nearly the same functional form as the Green's function (12),

$$C_s(v,v') = \frac{s2^s}{\sqrt{\pi}}\Gamma(s) e^{v'^2} R_s(-v_<) R_s(v_>). \tag{22}$$

Many other advantages of cusp kernels are described in reference [15] and also in reference [17], where it is shown that cusp kernels give excellent fits to laboratory measurements of atoms that are optically pumped under conditions similar to those of guidestar sodium atoms.

3. Publications

Activities receiving full or partial support from Grant AFOSR F49620-98-1-0127 have resulted in the 17 refereed publication, authored by the principal investigator and his colleagues, as listed below. The grant was also acknowledged in numerous abstracts, contributed and invited talks at scientific conferences.

1. (with K. Ishikawa, B. Patton, and Y.-Y. Jau) Spin Transfer from an Optically Pumped Alkali Vapor to a Solid, Phys. Rev. Letters **98**, 183004 (2007).

2. (with F. Gong and Y.-Y. Jau) Magnetic Resonance Reversals in Optically Pumped Alkali-Metal Vapor, Phys. Rev. A **75**, 053415 (2007).

3. (with B. Patton, K. Ishikawa and Y.-Y. Jau) Intrinsic Impurities in Glass Alkali-Vapor Cells, Phys. Rev. Letters **99**, 027601 (2007).

4. (with Y.-Y. Jau) Push-pull Laser-Atomic Oscillator, Phys. Rev. Letters, **99**, 223001 (2007).

5. (with F. Gong and Y.-Y. Jau) Nonlinear Pressure Shifts of Alkali-Metal Atoms in Inert Gases, Phys. Rev. Letters, **100**, 233002 (2007).

6. (with J. Ma, A. Kishinevski, Y.-Y. Jau and C. Reuter) Modification of glass cell walls by rubidium vapor, Phys. Rev. A, 79, 042905, (2009).

7. (with B. H. McGuyer and Y.-Y. Jau) Simple Method of Light-Shift Suppression in Optical Pumping Systems, Applied Physics Letters, **94**, 251l10 (2009).

8. (with T. G. Walker and Y.-Y. Jau) *Optically Pumped Atoms*, Wiley-VCH GmbH Verlag, Weinheim (2010).

9. (with T. Xia, S. W. Morgan and Y.-Y. Jau) Polarization and Hyperfine Transitions of Metastable ^{129}Xe in Discharge Cells, Phys. Rev. A, **81**, 033419, (2010).

10. (with S. W. Morgan) Optically Pumped Atoms with Velocity- and Spin-Changing Collisions at Low Gas Pressure, Phys. Rev. A, **81**, 042703, (2010).

11. (with N. Kostinski, B. Olsen, B. Marsland and B. McGuyer) Temperature-insensitive laser frequency locking near absorption lines, Rev. Sci. Instr. **82**, 033114 (2011).

12. (with I. Ishikawa, B. Patton, B.A. Olsen and Y.-Y. Jau) Transfer of spin angular momentum from Cs vapor to nearby Cs salts through laser induced spin currents, Phys. Rev. A **83**, 063410 (2011).

13. (with B. McGuyer, T. Xia, and Y.-Y. Jau) Hyperfine frequencies of ^{87}Rb and ^{133}Cs atoms in Xe gas, Physical Review A, **84**, 030501(R) (2011).

14. (with B.A. Olsen, B. Patton and Y.-Y. Jau) Optical pumping and spectroscopy of Cs vapor at high magnetic field, Phys. Rev. A **84**, 063410 (2011).

15. (with B. H. McGuyer, R. Marsland III and B. A. Olsen), Cusp Kernels for Velocity-Changing Collisions, Phys. Rev. Letters, **108**, 183202 (2012).

16. (with R. Marsland III, B. H. McGuyer and B. A. Olsen) Spin-Velocity Correlations of Optically Pumped Atoms, Phys. Rev. A **86**, 023404 (2012).

17. (with T. Bhamre, R. Marsland III, I. K. Kominis, and B. H. McGuyer) Collision Kernels from Velocity-Selective Optical Pumping with Magnetic Depolarization, Phys. Rev. A **87** 043412 (2013).

4. Personnel

The following personnel received full or partial support from Grant AFOSR F49620-94-1-0466:

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